

Self-trapping of light in an organic photorefractive glass

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We report the first observation, to our knowledge, of self-trapping of light as well as optically induced focusing-to-defocusing switching in an organic photorefractive glass, owing to the orientationally enhanced photorefractive nonlinearity of the material. © 2003 Optical Society of America

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In recent years spatial solitons have attracted considerable interest in nonlinear optics¹ as well as in other areas of physics, such as Bose–Einstein condensates. In this Letter we report the first experimental demonstration, to our knowledge, of optical spatial solitons in an organic photorefractive (PR) glass. Self-trapping of light, as well as optically induced focusing-to-defocusing switching, is observed in a PR monolithic glass based on a new type of nonlinear optical chromophore. We show that the orientational PR nonlinearity² that gives rise to spatial solitons can be switched from self-focusing to self-defocusing when the polarization of the optical beam is changed; however, it is not sensitive to the polarity of the bias (poling) field. These observations are in good agreement with the recent prediction of PR polymeric solitons^{3,4} based on low-cost but high-performance PR organic materials. With regard to potential applications we find that the speed of soliton formation depends on the intensity of the optical beam and the magnitude of the field, and it could be increased by synthetic modifications of the chromophores and by preorientation of the chromophores with the poling field.

The amorphous organic material used in our experiment is made from a mixture of two dicyanomethylene-dihydrofuran (DCDHF) chromophores (DCDHF-6 and DCDHF-6-C7M) at a 1:1 weight ratio,^{5,6} sensitized with 0.5 wt.% of the charge generator buckminsterfullerene (C₆₀). The DCDHF chromophores provide a poled array of dipolar molecules with large polarizability anisotropy, which makes them highly birefringent. It has been shown that mixing DCDHF-6 and DCDHF-6-C7M chromophores leads to the formation of stable glass ($T_g = 23^\circ\text{C}$) that resists crystallization for at least 2 years.^{5,6} The absorption spectra of the two DCDHF chromophores are similar because of their structural similarity, and they have an absorption maximum at ~ 490 nm. In previous experiments high two-beam-coupling gain was achieved in this

material at a wavelength of 676 nm. In contrast to the wave-mixing geometry, our soliton experiment requires that a light beam propagate in the plane of the thin-film material over a distance of the sample length (typically 2.5 mm or more). For such a long propagation distance, a higher wavelength of 780 nm is used to avoid excessive absorption by the chromophores while maintaining a nonzero absorption from C₆₀ for photogeneration of charges.

In our experiment a 780-nm laser diode, along with an anamorphic prism pair and collimating lenses, is used to provide a nearly circular beam. A half-wave plate rotates the polarization of the beam when necessary. The collimated beam is then focused with a cylindrical lens onto the input face of a 120- μm -thick sample of the PR organic glass. To allow optical propagation over the 2.5-mm distance in the plane of the sample, the front and back edges of the sample are covered with a piece of glass to create nearly flat input and output surfaces. As shown in Fig. 1, the beam propagates along the z direction through the film while a dc electric field is applied between indium tin oxide electrodes along the x direction. Behind the sample a CCD camera is used along with an imaging lens to monitor the beam profiles directly at the input and output faces of the organic material. With such a setup, self-trapping of light is observed when the

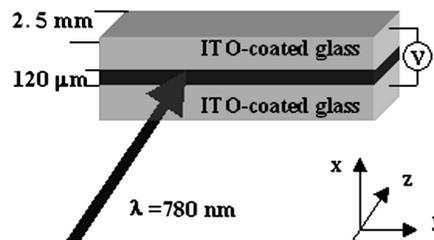


Fig. 1. Sketch of the experimental geometry showing the sample orientation. ITO, indium tin oxide.

beam is polarized in the y direction (i.e., perpendicular to the bias field).

Typical experimental results are presented in Fig. 2. The power of the laser beam before the sample was set at ~ 24 mW, and the beam was focused to $12\ \mu\text{m}$ FWHM at the input face of the sample [Fig. 2(a)]. Without the applied electric field the beam diffracted to $\sim 55\ \mu\text{m}$ after 2.5 mm of linear propagation [Fig. 2(b)]. With the applied field, self-focusing was observed when the beam was polarized perpendicular to the bias field, and the strength of the focusing increased gradually. Furthermore, at a dc field of $16\ \text{V}/\mu\text{m}$, self-trapping of the beam into an optical soliton was realized, and the beam reached its original size in ~ 160 s [Fig. 2(c)]. The soliton was then stable against small power fluctuations.⁷ After 1 h, a noticeable increase in the beam width was observed. Additional background illumination seems to favor steady-state soliton formation, as is the case for PR screening solitons.⁸ The experiment was repeated in a few other samples made of the same composite material, and similar results were reproduced.

The self-focusing effect does not depend on the polarity of the dc field, because it was also observed when the polarity of the dc field was reversed. This is in contrast with the optical nonlinearity in most noncentrosymmetric inorganic PR crystals, for which the index change induced by the space-charge field is determined by the linear electro-optic (EO) effect. In fact, in an inorganic PR crystal such as strontium barium niobate, reversing the polarity of the bias field would change the crystal from self-focusing to self-defocusing, as was demonstrated with PR screening solitons.⁸ In our organic PR glass, however, the nonlinearity does depend on the polarization of the beam itself. When the polarization was switched to be parallel with the applied field, self-defocusing was observed instead of self-focusing.⁷ In this case the beam width increased to $\sim 80\ \mu\text{m}$ after a field of $16\ \text{V}/\mu\text{m}$ was applied [Fig. 2(d)]. This demonstration represents a convenient way to optically induce switching from self-focusing to self-defocusing. (Previously, such switching was realized in photovoltaic PR crystals when the nonlinearity was fine-tuned with additional optical illumination⁹).

The observed effect can be explained well by the theory of orientational PR nonlinearity.² The refractive-index change Δn caused by reorientation of the chromophores depends on both the resulting birefringence (BR) and the EO effect. With a dc electric field (E_{dc}) applied in the x direction, one can find the change of the refractive index for either x - or y -polarized light beams under certain approximations²⁻⁴:

$$\begin{aligned}\Delta(n^2)_x &= \Delta(n^2)_x^{\text{BR}} + \Delta(n^2)_x^{\text{EO}} \\ &\cong (C_x^{\text{BR}} + C_x^{\text{EO}})E_{\text{dc}}^2 \cong C_x E_{\text{dc}}^2, \\ \Delta(n^2)_y &= \Delta(n^2)_y^{\text{BR}} + \Delta(n^2)_y^{\text{EO}} \\ &\cong (C_y^{\text{BR}} + C_y^{\text{EO}})E_{\text{dc}}^2 \cong C_y E_{\text{dc}}^2,\end{aligned}\quad (1)$$

where $C_x^{\text{BR}} = (2/45)(4\pi N)\Delta\alpha(\mu/k_B T)^2$, $C_x^{\text{EO}} = (1/5)(4\pi N)\beta_{333}(\mu/k_B T)$, and N is the number density

of the chromophores. $\Delta\alpha = \alpha_{\parallel} - \alpha_{\perp}$, which represents the polarizability anisotropy, with $\alpha_{\parallel} = \alpha_{33}$ and $\alpha_{\perp} = \alpha_{11} = \alpha_{22}$ for uniaxial chromophores. Here μ is the permanent dipole moment of the molecule, k_B is the Boltzmann's constant, T is the absolute temperature, and β_{333} is the dominant term of hyperpolarizability for a rodlike molecule. In most low-glass-transition organic PR materials, such as the one used in our experiment, BR dominates the PR nonlinearity. Since $C_x^{\text{BR}} = -C_x^{\text{EO}}/2$, neglecting the EO term from relations (1), one can obtain $\Delta(n^2)_x \cong C_x^{\text{BR}}E_{\text{dc}}^2$ and $\Delta(n^2)_y \cong -1/2 C_x^{\text{BR}}E_{\text{dc}}^2$. This suggests that in such a biased material an x -polarized light beam experiences a positive Δn due to enhanced BR, whereas a y -polarized light beam experiences a negative Δn [$\Delta\alpha = (\alpha_{\parallel} - \alpha_{\perp}) > 0$ is usually the case]. When a nonuniform Gaussian-like beam is launched into the sample, it creates a space-charge field that partially screens the electric field E_{dc} in the illuminated region, which results in a difference in refractive-index change between the illuminated and nonilluminated parts of the sample. Intuitively, this explains why the soliton generation requires a y -polarized beam and the self-defocusing requires an x -polarized beam.⁴ As seen from relations (1), reversing the polarity of the bias field here does not affect Δn because of a quadratic dependence on the field. However, simply by rotating the polarization, a transition from self-focusing to self-defocusing could be realized as demonstrated in our experiment.

The self-focusing experiment in a given sample was performed at various incident-beam power levels and at various bias field strengths. Figure 3 shows typical results of varying the bias field while all other conditions were kept the same. Between successive experiments the electric field and the laser beam were turned off, and the sample was washed out with white

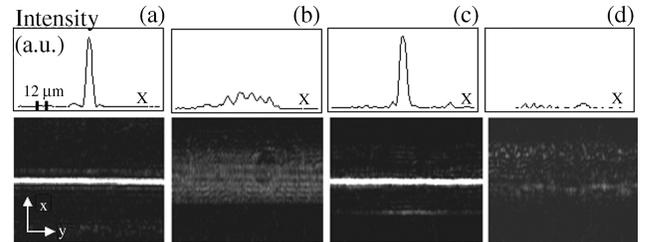


Fig. 2. Self-trapping and self-defocusing of a light beam in a PR organic glass.⁷ Shown are (top) intensity profiles along the x direction and (bottom) transverse patterns taken at (a) the sample input and (b–d) output. (b) Normal diffraction with no nonlinearity. (c) Self-trapping. (d) Self-defocusing with nonlinearity.

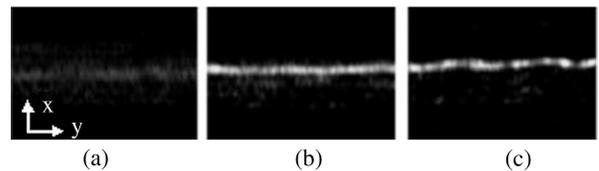


Fig. 3. Output intensity patterns of a stripe beam at various values of the bias field. From left to right the field is 8, 16, and $25\ \text{V}/\mu\text{m}$.

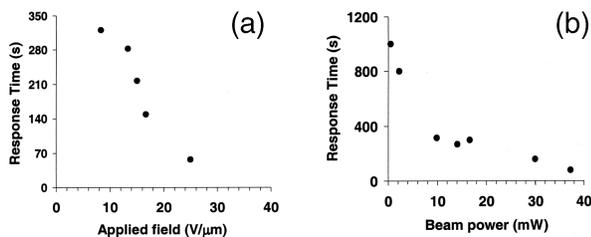


Fig. 4. Response time for the material to reach maximum nonlinearity as a function of (a) applied field at a fixed power of 29 mW and (b) beam power at a fixed bias field of 16 V/μm.

light (to destroy any nonuniform internal charge distribution) until the residual index change disappeared completely. The photographs were taken when maximum self-focusing was reached for each value of the bias field. From left to right the field was increased gradually. At a fixed beam power it appears that there is a critical value of the electric field that favors soliton formation. If the field is too low, the beam is only partially focused and not strong enough to balance diffraction [Fig. 3(a)]. On the other hand, if the field is too high, the nonlinearity is so strong that the beam breaks up into filaments as a result of transverse modulation instability⁸ [Fig. 3(c)]. A soliton forms only at appropriate conditions, which are better described by the so-called soliton existence curve.^{4,8}

The time it takes for the beam to reach maximum self-focusing or for the material to reach maximum change of the refractive index depends on the intensity of the optical beam as well as the strength of the bias field. To estimate this response time in our experiment, images were taken at a fixed rate, and the time it takes for the beam to reach its peak intensity due to self-focusing was found in each measurement. When the beam power incident on the sample was kept at the same level, self-focusing occurred faster at a higher bias field, as shown in Fig. 4(a). Likewise, when the applied field was kept constant, self-focusing occurred faster at a higher beam power [Fig. 4(b)]. In particular, at an electric field of 16 V/μm the response time decreased by more than an order of magnitude (from 1000 to 80 s) as the incident beam power was increased from 0.5 to 37 mW [Fig. 4(b)]. We performed all measurements described so far by first launching the light beam into the sample, then applying the bias field. For comparison we reversed the experimental sequence; the sample was prepoled first by application of an electric field for more than 30 min, then, with the poling field on, the beam was launched into the sample. We found that self-trapping of the beam occurred noticeably faster in the prepoled sample. In yet another experiment with a sample sensitized with 2.0 wt. % of C₆₀ it took less than 10 s for a soliton to form under the same experimental conditions as for Fig. 2. For applications, improvement of the soliton

formation speed will be the subject of future research. For instance, since orientational dynamics in organic glasses strongly depends on the temperature relative to the glass transition temperature (T_g), fine-tuning T_g of the materials could dramatically improve the speed. It is expected that a response time of 1 s or less (as opposed to 100 s or more) can be achieved with fine-tuning of the temperature to a few degrees above T_g . This could be accomplished by synthetic modifications of the DCDHF chromophores as well as by mixing various DCDHF derivatives in appropriate concentrations.⁶

In summary, we have demonstrated for the first time to our knowledge self-trapping of light in an organic PR glass, along with an effective approach for switching between self-focusing and self-defocusing. Applications proposed with solitons in inorganic PR crystals¹⁰ and in liquid crystals¹¹ might also be realized in organic PR glasses, while taking advantage of such materials for their low cost and structure flexibility compared with inorganic crystals and for their amenable absorption and scattering compared with liquid crystals. For instance, it might be possible to develop directional couplers and efficient nonlinear frequency converters in soliton-induced waveguides based on organic materials.^{10,12}

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